Structure of Local Spin Excitations in a Geometrically Frustrated Antiferromagnet

For many crystalline magnetic materials, only a long range ordered spin configuration can satisfy all near neighbor spin interactions. Such systems generally display a finite temperature transition to a broken symmetry phase with long-range magnetic order. However, on certain lattices with weak connectivity and a triangular motif, short-range interactions can be satisfied without long-range order [1].

To explore this possibility we examined magnetic order and fluctuation in $ZnCr_2O_4$. The B-site of this spinel lattice is solely occupied by spin-3/2 Cr^{3+}, and this leads to a magnet with dominant nearest neighbor interactions on the lattice of corner-sharing tetrahedra shown in Fig. 1.

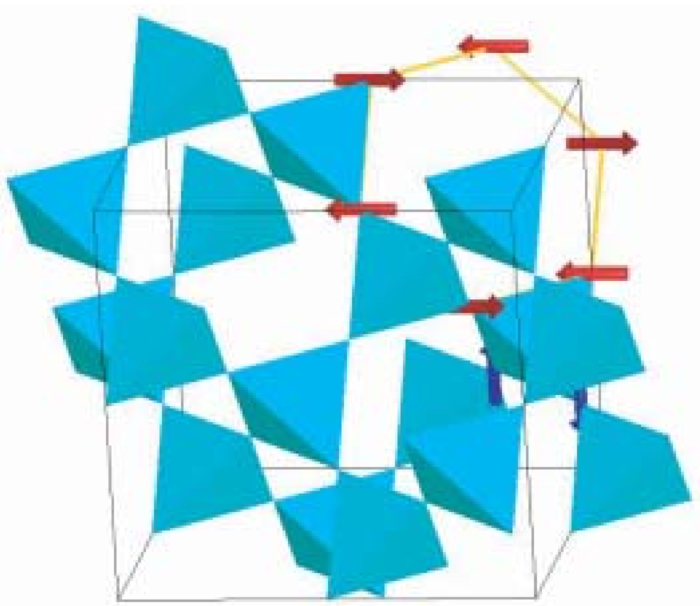

FIGURE 1. Lattice of corner-sharing tetrahedral formed by the octahedrally coordinated B sites of the spinel AB_2O_4. Black lines represent a structural unit cell containing eight tetrahedra. Vertices are populated by antiferromagnetically interacting spin-3/2 degrees of freedom in $ZnCr_2O_4$. Yellow line and six red spins represent a hexagonal loop of antiferromagnetically aligned spins.

Exchange interactions in this structure are satisfied at the mean field classical level when every tetrahedron carries zero net magnetization. Not only are there many ways that a tetrahedron can have no moment, but there are also many ways to organize such non-magnetic tetrahedra on the corner-sharing lattice. The result is a “geometrically frustrated” spin system with many ways to satisfy interactions without long-range order.

Analysis of specific spin models on the B-site spinel (or pyrochlore) lattice indicates that spin-1/2 and spin-∞ models have short-range order down to $T = 0$ [2], while long-range order is induced by quantum fluctuations for an unknown intermediate range of spin values. Experiments indicate that spin-3/2 $ZnCr_2O_4$ is “close” to the quantum critical point that separates the low spin quantum-disordered phase from the intermediate spin long-range ordered phase. Specifically, the relaxation rate for magnetic excitations, Γ, follows a power-law that extrapolates to zero as T approaches 0, indicating quantum criticality [3]. This state of affairs, however, does not persist to the lowest temperatures. Instead, at $T_N = 12.5$ K a first order structural transition from the cubic cooperative paramagnet to tetragonal Néel order intervenes.

Figure 2 shows that a gapless continuum of magnetic scattering above T_N is pushed into a local spin resonance at $\hbar\omega \approx 4.5$ meV $\approx J$ with remarkably little dispersion throughout the Brillouin zone. The result is analogous to the spin-Peierls transition of the uniform spin-1/2 chain, where quantum critical fluctuations are pushed into a finite energy singlet-triplet transition through structural dimerization. Our

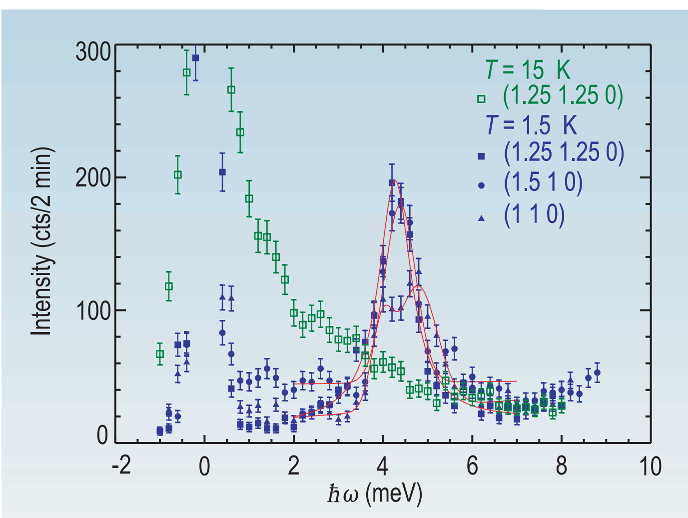

FIGURE 2. Constant-Q scans above and below T_N = 12.5 K. In the cooperative paramagnetic phase above T_N magnetic excitations form a continuum. Below T_N the spectrum changes dramatically with a nearly dispersionless excitation near 4.5 meV appearing abruptly. The red curves are to guide the eye.

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recent synchrotron x-ray and neutron powder experiments indicate that deformation of tetrahedra does indeed occur for $ZnCr_2O_4$. However, the structural changes push the system to long range order rather than to quantum disorder, as indicated by the magnetic Bragg peaks and spin wave excitations.

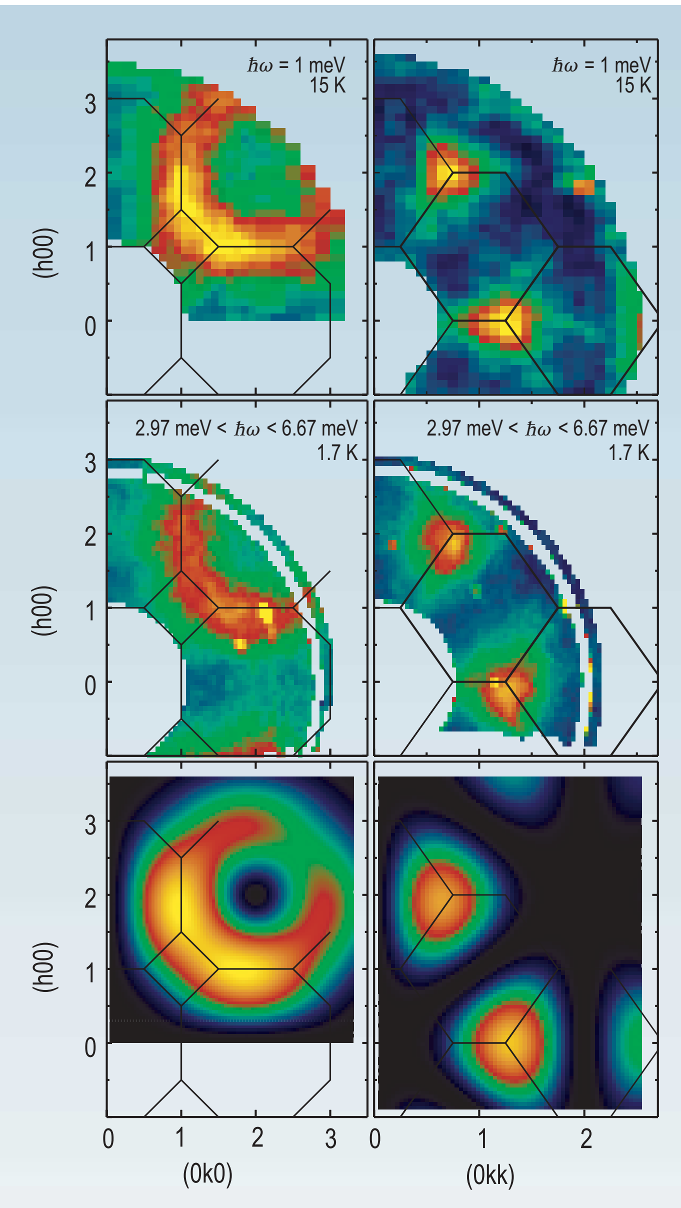

FIGURE 3. Color images of inelastic neutron scattering intensities in the (hk0) and (hkk) symmetry planes. From top to bottom are shown data from below and above T_N, and the structure factor for the hexagon mode proposed by O. Tchernyshyov. T = 1.7 K data were taken integrating over the energy range $\hbar\omega$ from 3.0 meV to 6.7 meV. Data in the paramagnetic phase (T = 15 K) were taken at $\hbar\omega$ = 1 meV.

Our single crystal experiment [4] also enabled unique insight into the local structure of spin-fluctuations in geometrically frustrated systems. Figure 3 shows the $\boldsymbol{Q}$-dependence of low energy magnetic scattering in two high symmetry planes above and below T_N. While the spectrum for spin fluctuations changes dramatically at the first order phase transition, the structure factor clearly does not. Also shown in the figure is the structure factor for six spins of <111> type kagomé hexagons precessing with π phase shift between neighbors. The proposal by O. Tchernyshyov *et al.*, [5] that these are the dominant low energy spin fluctuations for spins on the B-site spinel lattice is clearly borne out by the data.

The present data for $ZnCr_2O_4$ show that geometrically frustrated lattices have composite low energy degrees of freedom analogous to rigid unit modes in certain open framework lattice structures. To better understand the unusual type of phase transition that occurs in this system, it must be determined what defines the 4.5 meV energy scale for hexagon excitation in the ordered phase. Do quantum fluctuations play a significant role or does the broken symmetry between exchange interactions within tetrahedra induce the resonance? The answer to this question is now being pursued through an accurate determination of the complex low temperature lattice and magnetic structure.

References

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